



Effects of Siberian forest fires on air quality in East Asia during May 2003 and its climate implication

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ABSTRACT

In May 2003, intense forest fires occurred over Siberia, which were the largest fires in the past decade. In order to quantify the effects of these fires on regional air quality in East Asia, we used a global chemical transport model (CTM) with a biomass burning emission inventory constrained by satellite. Our focus was mainly on the enhancements of the ozone and aerosol concentrations due to these fires over East Asia. We first evaluated the model extensively by comparing the simulated and the observed ozone and aerosol concentrations at the EANET sites and found that the simulation reproduced the observed variability of those species. However, some discrepancies were found in the model when compared with the MODIS AOD observations. We tested the sensitivity of the model AOD to different injection heights of fire emissions and found that the model with an injection height of 4.5 km was in better agreement with the observations. We then used our model results to quantify the influences of Siberian forest fires on ozone and aerosols concentrations which were computed using the differences between the simulations with and without Siberian forest fire emissions. The peak increases in the surface PM_{10} and ozone concentrations were up to $90 \mu g m^{-3}$ and 33 ppbv, respectively, over Siberia. In the downwind regions, the increases ranged from 5 to $30 \mu g m^{-3}$ and from 3 to 20 ppbv for PM_{10} and ozone concentrations, respectively, having an important implication for air quality over East Asia. Finally, we computed the radiative forcing of aerosols and ozone from the Siberian forest fires as a measure of climate impact. Siberian forest fires were found to act mainly as a cooling agent resulting in a negative radiative forcing of $-5.8 W m^{-2}$ at the surface over East Asia. The value at the TOA was $-1.5 W m^{-2}$, indicating that a considerable absorption of radiation occurred in the atmosphere. This result implies that the Siberian forest fires may affect the regional climate over East Asia by intensifying atmospheric stability.

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1. Introduction

Forest fires are one of the major sources of CO, volatile organic compounds (VOCs), and nitrogen oxides $NO_x \equiv NO + NO_2$ and thus have a significant effect on tropospheric ozone (Crutzen et al., 1979; DeBell et al., 2004). They also

release high concentrations of aerosols into the atmosphere and result in a severe visibility degradation and harmful effects on human health (Bowman and Johnston, 2005; Park et al., 2006, 2007; In et al., 2007). The ozone precursors and aerosols from forest fires are transported over long distances, affecting air quality in downstream regions (Wotawa and Trainer, 2000; Forster et al., 2001; Wotawa et al., 2001; McKeen et al., 2002; Bertschi et al., 2004; Colarco et al., 2004; Jaffe et al., 2004; Park et al., 2007; Spracklen et al., 2007). In addition, ozone and aerosols have important climatic implications because of their

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effects on the earth-atmosphere radiative system (Li et al., 2001; Pfister et al., 2008).

In May 2003, intense forest fires occurred in Siberia, which were recorded as one of the largest fires in Siberia in the past decade. Resulting smoke plumes and associated high aerosol optical depth (AOD) were measured by satellite (Lee et al., 2005). Surface observations also showed a significant enhancement in aerosol concentrations due to Siberian fires even in the far downwind regions (Bertschi and Jaffe, 2005; Lee et al., 2005; Nedelec et al., 2005; Lapina et al., 2006; Kaneyasu et al., 2007). The effects of those fires on air quality have been quantified in the previous studies for ozone in North America (Jaffe et al., 2004) and for column aerosols north of 75°N (Generoso et al., 2007). Because of the close proximity to the locations of the fires, East Asia must be heavily affected by the smoke plumes from these fires. However, there has been no extensive quantification of the effects of these fires on regional air quality on a daily and monthly basis in East Asia.

In this study, we use a global chemical transport model (CTM) to quantify the effects of the Siberian forest fires on regional air quality over East Asia in May 2003. Our focuses are primarily on the regional enhancements of ozone and aerosols concentrations due to the Siberian forest fires in May 2003. Although forest fires that occurred over Siberia in June 2003 were large compared with those of other years, their impacts over East Asia were much less than those in May because of 1) by a factor of two smaller emissions relative to those of May (van der Werf et al., 2006) and 2) the onset of Asian summer monsoon which accompanied large precipitation and likely scavenged the aerosols (Yihui and Chan, 2005).

The intense thermal energy which accompanies forest fires causes a rapid vertical transport of the smoke plumes into the free troposphere well above the planetary boundary layer (PBL) (Lavoué et al., 2000; Colarco et al., 2004; Fromm et al., 2005; Mazzoni et al., 2007). Recent studies have shown that smoke aerosols from forest fires reached the upper troposphere (Fromm and Servranckx, 2003; Jost et al., 2004; Turquety et al., 2007). The injection height of the forest fire emissions is one of the critical factors in determining the spatial range of fire plume transport and hence the air quality in the downwind regions. However, it has not been well constrained in the model. We also examine the sensitivity of the model to different injection heights of fire emissions by comparing the simulated results with the observations at the surface network and from the satellite measurements.

High aerosol and ozone concentrations from forest fires may perturb the radiative balance and cause a change in the regional climate (Duncan et al., 2003; Liu, 2005; Pfister et al., 2008). This issue will be of increasing importance in the future since the number of forest fires is expected to increase with increasing temperature in the warming climate (Stocks et al., 1998; Westerling et al., 2006; Soja et al., 2007; Malevsky-Malevich et al., 2008). As a measure of the climatic impact of forest fires, we use our model to estimate the radiative forcing of the aerosols and ozone from the Siberian fires, which has implications for the regional climate in East Asia.

2. Model simulations

We use the GEOS-Chem chemical transport model (version 7.04) to conduct a fully coupled oxidant-aerosol simulation (Park et al., 2006). The GEOS-Chem model uses the assimilated meteorological data from the Goddard Earth Observing System (GEOS-4) of the NASA Global Modeling and Assimilation Office (GMAO). The data include winds, convective mass fluxes, temperature, clouds, and precipitation at 6-h frequencies (3-h frequencies for surface quantities and mixing depths) with a horizontal resolution of $1^\circ \times 1^\circ$ and 55 hybrid pressure-sigma levels up to 0.01 hPa. We degrade these meteorological fields to a horizontal resolution of $2^\circ \times 2.5^\circ$ and 30 vertical levels for computational expediency.

The GEOS-Chem includes more than 80 species and 300 reactions for a detailed ozone- NO_x -hydrocarbon chemistry coupled with aerosol chemistry. Ozone simulations were evaluated extensively in the troposphere (Bey et al., 2001; Fiore et al., 2002, 2003; Hudman et al., 2004). The aerosol simulation includes H_2SO_4 - HNO_3 - NH_3 aerosol thermodynamics, primary organic carbon (OC) and elemental carbon (EC), secondary organic aerosol (SOA), soil dust, and sea salt (Park et al., 2003, 2004, 2005; Heald et al., 2005; Alexander et al., 2005; Fairlie et al., 2007). SOA formation follows the scheme of Chung and Seinfeld (2002). Mobilization, transport, and deposition processes of soil dust aerosols with 4 size bins were simulated using the methods described by Fairlie et al. (2007) with the Dust Entrainment and Deposition (DEAD) scheme of Zender et al. (2003a,b). The sea salt emission is computed as a function of dry particle size and local 10 m wind speed following the empirical formula from Monahan et al. (1986) (Alexander et al., 2005).

All inorganic aerosols, primary OC and EC, and SOA aerosols are considered as fine aerosols smaller than $2.5 \mu\text{m}$ in diameters and are included in $\text{PM}_{2.5}$ and PM_{10} mass concentrations in the model. Soil dust and sea salt aerosols in the model are partitioned into $\text{PM}_{2.5}$ and PM_{10} based on their sizes. For the AOD computation using aerosol dry mass concentrations, particle growth with increased relative humidity is taken into account by applying different hygroscopic growth factors to all hydrophilic species using local relative humidity conditions (Martin et al., 2003; Liu et al., 2004). We also account for the non-carbon mass attached to OC aerosol by applying a scaling factor of 2.0 to the primary OC aerosol concentration for the $\text{PM}_{2.5}$, PM_{10} , and AOD computations in the model (Turpin and Lim, 2001). We refer to the sum of OC with the non-carbon mass and SOA as organic carbon mass (OMC) hereafter.

We use the 1999–2000 global inventories of the anthropogenic emissions of NO_x , CO, VOCs, SO_x , NH_3 , and primary aerosols as discussed in Park et al. (2006). The Asian emissions of NO_x , CO, VOCs, SO_x , and NH_3 defined for 60°E – 158°E and 13°S – 54°N are 9.1 Tg N y^{-1} , 259.8 Tg y^{-1} , 24.9 Tg C y^{-1} , 18.9 Tg S y^{-1} , and 21.8 Tg N y^{-1} , respectively. The anthropogenic emissions of primary OC and EC are from Bond et al. (2004); Asian anthropogenic OC and EC emissions are 4.9 Tg C y^{-1} , and 2.6 Tg C y^{-1} , respectively. Other emissions included those from volcanoes, lightning, the biosphere (terrestrial and marine), and biomass

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